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On the determination of the electron-capture decay energies of Tl^{201} and Tl^{202}

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With 2 figures in the text

ABSTRACT

The decay energy of Tl^{201} has been calculated by deducing the ratio between the probabilities of L_I- and K-electron capture to the 167 keV state in Hg^{201} from the number of coincidences between the 167 keV gamma ray and the K X-rays. The value of the total decay energy was found to be $405\,^{+70}_{-40}$ keV. In a similar way the total decay energy of Tl^{202}, calculated from the number of coincidences between the 965 keV gamma ray and the K X-rays, was found to be 1110 ± 20 keV.

1. Introduction

In the determination of the electron-capture (hereafter referred to as E.C.) decay energy one has to use indirect methods, as the total disintegration energy during the E.C. process is carried away by the neutrino, on which it is hardly possible to make direct measurements. In the case of E.C. isotopes where no gamma rays are emitted in the decay process, one can obtain a reliable value of the decay energy from the measurement of the internal bremsstrahlung spectrum associated with the E.C. decay. However, when gamma rays are emitted in the decay process, it is difficult to measure the weak internal bremsstrahlung spectrum. In such cases a reliable value of the decay energy can be obtained, if one can determine the ratio between the probabilities of L_I-electron capture and K-electron capture, P_{L_I}/P_K, to the highest excited state of the daughter nucleus [1].

In the case of Tl^{201} and Tl^{202}, the ratio P_{L_I}/P_K for decay to the highest excited state can be obtained by studying coincidences between the most energetic gamma ray and the K X-rays by a conventional coincidence set-up using two NaI (Tl) detectors.

2. Source preparation

The Tl^{201} and Tl^{202} activities were produced by milking thallium from active lead produced by bombarding thallium metal with about 40 MeV protons in the synchrocyclotron of the Gustaf Werner Institute in Uppsala. The chemical method of separating lead from irradiated thallium was the same as described in [2]. The grown-in thallium activity was extracted with ether saturated with 6 N HCl. In the case of Tl^{202} sources, the active lead was separated immediately after the bombardment and the grown-in thallium activity was milked after about 12 hours. The Tl^{202} was studied after about one month, when all short-lived activities had decayed away. In the case of Tl^{201}, the active lead was separated from the irradiated thallium after about 10 hours, and the grown-in thallium activity was extracted 8 hours later. The Tl^{201} studies were started after about one week, when Tl^{200} had decayed away.

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3. Experimental procedure and results

The scintillation spectra of the thallium sources were studied with a conventional scintillation spectrometer using a $1\frac{1}{2}'' \times 1''$ or a $3'' \times 3''$ NaI (Tl) crystal. A Hutchinson-Scarrott type multichannel analyser was used for pulse-height analysis. The coincidences between gamma rays and K X-rays were studied in a coincidence scintillation spectrometer using single channel analysers in both channels [3]. In order to maintain 100 per cent coincidence efficiency over the entire K X-ray peak, a resolving time of 0.15 μsec was used. The ratio of the number of coincidences between the K X-ray and the gamma ray, $I_{\gamma x}$, to the number of gamma rays, I_γ, is given by

$$I_{\gamma x}/I_\gamma = (N_K/N)\,\omega_K\, S\, e^{-\mu_x t}\,\varepsilon_x = P_K\,\omega_K\, S\, e^{-\mu_x t}\,\varepsilon_x\,, \tag{1}$$

where N_K is the number of K-electron capture transitions leading to the excited state and N is the total number of capture transitions leading to the same state $(N_K/N = P_K)$, ω_K is the K-fluorescence yield, S is the solid angle subtended by the source at the X-ray detector crystal, ε_x is the efficiency of the crystal for the K X-ray and $e^{-\mu_x t}$ is a transmission factor for the cover of the crystal. In calculating $e^{-\mu_x t}$, the angular variation of the transmission through the cover of the crystal was taken into account.

Tl²⁰¹

A detailed investigation of the decay of 3 day Tl^{201} has recently been carried out at this laboratory [4], and the results are in agreement with the decay scheme presented in the inset of Fig. 1.

The scintillation spectrogram of Tl^{201} is given in Fig. 1. One can see the escape and full-energy peaks of the K X-rays, and the photopeaks of the 135 and 167 keV gamma rays. The low intensity peaks at 368 and 439 keV are due to Tl^{200} and Tl^{202}, respectively. The Compton distribution from the 368 and 439 keV gamma rays gave a contribution of less than one per cent to the photopeak of the 167 keV gamma ray. In the coincidence study of the 167 keV gamma ray and the K X-rays, the pulses corresponding to the higher part of the 167 keV photopeak were fed into the gate in order to avoid contribution from the 135 keV gamma ray. The results of the coincidence measurements carried out with different distances between the source and the X-ray detector are summarized in Table 1. The value 0.954 of the K-fluorescence yield ω_K of mercury has been used [5]. As a mean value from the three measurements P_K is found to be 0.70 ± 0.04. Using the $(P_M + P_N + \cdots)/P_L$ ratio given in ref. [5] and the theoretical results of Brysk and Rose [6] for non-unique, first forbidden transitions, one can write

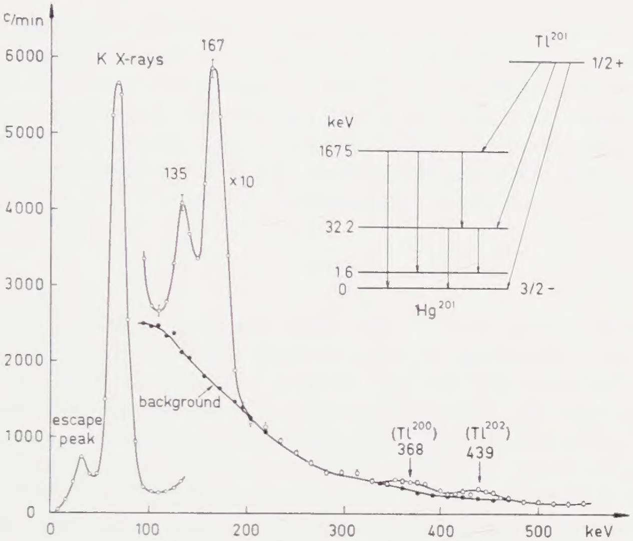

Fig. 1. Scintillation spectrum of Tl^{201} taken with a $1\frac{1}{2}'' \times 1''$ NaI (Tl) crystal. The inset shows the decay scheme of Tl^{201}.

Table 1.

S	ε_x	$e^{-\mu_x t}$	$I_{\gamma x}$	I_γ	P_K
0.0635	0.975	0.900	222.8	6195.5	0.677
0.0343	0.980	0.905	158.8	7703.0	0.710
0.0140	0.990	0.910	48.16	5634.0	0.710

$$\begin{aligned} &P_{L_{II}}/P_{L_I} = 0.080; \quad P_{L_{III}} = 0 \\ &(P_M + P_N + \cdots)/P_L = 0.268 \\ &P_{L_I}/P_K = 0.151 \left(\frac{E_0 - 14.84}{E_0 - 83.11}\right)^2 \end{aligned} \tag{2}$$

Substituting the experimental value of P_K, the decay energy to the 167 keV level is calculated to be $238\ {}^{+70}_{-40}$ keV. The total transition energy is then found to be

$$E_d\,(Tl^{201}) = 405\ {}^{+70}_{-40}\ \text{keV}$$

The ground state of Tl^{201} is assigned $1/2+$ [7]. Since the multipolarity of the 167 keV transition is proved to be $M1$ [4], and since the ground state of Hg^{201} is assigned $3/2-$ [8], the 167 keV state of Hg^{201} can be assigned $5/2-$, $3/2-$ or $1/2-$. However, the calculations above have been made on the basis of assignments $3/2-$ and $1/2-$, only (non-unique, first forbidden transition). This seems justified according to the following consideration. Using the above value of E_d and the fact that about 25 per cent of the K-capture transitions feed the 167 keV state [4], one finds a $\log ft$ value of about 6 for the K-capture transition to this state.

This is thus consistent with a non-unique, first forbidden transition. On the other hand, if one assumes the 167 keV state to be 5/2 –, the total decay energy, using the theoretical results of Brysk and Rose [6], comes out to be $590\ ^{+140}_{-80}$ keV. From this value one finds that $\log ft$ is about 6.5 for the K-capture transition to the 167 keV state. This, however, also indicates a non-unique, first forbidden transition, in contradiction with the assumption of a 5/2 – assignment for the 167 keV state. Thus the 1/2 – or 3/2 – assignments of the 167 keV state are favoured.

Tl^{202}

The total decay energy of Tl^{202} has been found to be $600\ ^{+5}_{-7}$ keV by deducing P_L/P_K for the E.C. transition to the 439 keV state of Hg^{202} from the number of coincidences between the 439 keV gamma ray and the K X-ray, measured by a summing technique [9]. Later, Hamers *et al.* [10] determined the total decay energy of Tl^{202} to be 599 ± 5 keV. However, from the beta-decay systematics, one expects the decay energy of Tl^{202} to be about 1.4 MeV (interpolation between the values 2.46 and 0.35 MeV for the decay energies of Tl^{200} and Tl^{204}, respectively).

Recently it was reported by Born *et al.* [11] that, apart from the well-known 439 keV gamma ray, two low intensity gamma rays of 965 ± 8 and 523 ± 2 keV are also associated with the E.C. decay of Tl^{202}, the latter gamma ray being in coincidence with the 439 keV gamma ray. Thus it appeared that the decay energy of Tl^{202} must be more than 965 keV (see also ref. [12]). Also from the earlier results [9], after applying a correction for captures from higher shells (M, N, . . .) and an accurate absorption correction for the K X-rays in the cover of the crystal, it appeared that the decay energy of Tl^{202} is about 1 MeV.

It can be seen from a plot of P_{L_I}/P_K versus E_0 (decay energy) that the calculation of the decay energy by deducing P_{L_I}/P_K for the E.C. transition to the 439 keV state from the number of coincidences between the 439 keV gamma ray and K X-rays will involve large uncertainties (cf. formulae (2)). It seemed worth while to reinvestigate the decay of Tl^{202} and calculate the decay energy of Tl^{202} by deducing P_{L_I}/P_K to the 965 keV state from the number of coincidences between the 965 keV gamma ray and the K X-rays. While this work was in progress, the decay energy of Tl^{202} was reported to be $1100\ ^{+400}_{-175}$ keV [13], obtained from the number of coincidences between the 439 keV gamma ray and the K X-rays. A recent similar study of Tl^{202} by Jha *et al.* [14] also confirms the results of references [11] and [13].

Tl^{202} was investigated, after the short-lived isotopes had decayed away, by use of a $3'' \times 3''$ NaI (Tl) crystal. In order to reduce the intensity of the 439 keV gamma ray and to avoid overloading the electronic equipment, a 1 cm lead absorber was put between the source and the crystal. The scintillation spectrogram of Tl^{202} is shown in Fig. 2, where one can see the 965 keV gamma peak clearly. The intensity of the 965 keV gamma ray has been estimated to be (0.10 ± 0.03) % of the 439 keV gamma ray.

From the gamma-gamma coincidence experiments it was concluded that a gamma ray of about 520 keV is in coincidence with the 439 keV gamma ray. This is in agreement with the results reported in refs. [11] and [14]. Thus it appears that the 965 keV state is the second excited state of Hg^{202}. The decay scheme of Tl^{202}, based on these measurements, is shown in the inset of Fig. 2.

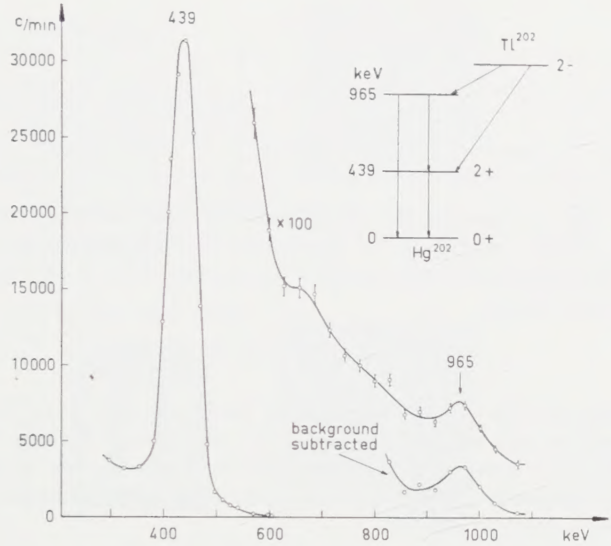

Fig. 2. Scintillation spectrum of Tl^{202} taken with a 1 cm lead absorber on the top of the 3″ × 3″ NaI (Tl) crystal. The inset shows the decay scheme of Tl^{202}.

In order to determine the decay energy of Tl^{202}, the following experiments were carried out. Firstly, the coincidence rate between the 439 keV gamma ray and the K X-rays was measured. As a result of these experiments, the value of P_K was found to be 0.79 ± 0.03. Secondly, the number of coincidences between the 965 keV gamma ray and the K X-rays was measured taking into consideration the following details:

1) 1 cm lead was put between the source and the gamma detector to reduce the random pile-up peak at about 880 keV.
2) In order to keep the ratio between true and random coincidences high (~ 10), a resolving time (2τ) of 50 nsec was used.
3) The high energy portion of the 965 keV photopeak was taken in the gate to avoid any contribution from the random sum-up peak due to the 439 keV gamma rays.
4) Only a narrow channel on the K X-ray peak was taken in order to avoid variations of the coincidence efficiency.
5) $(I_{\gamma x}/I_\gamma)_{965}$ was compared with $(I_{\gamma x}/I_\gamma)_{439}$ for the same setting.
6) The coincidence rate was also checked after covering the lead absorber with 1 mm Cd, in order to find out if some of the coincidences were due to X-rays produced in the lead.
7) The coincidence rate was also checked by taking the lower portion of the photopeak and a small portion of the Compton continuum due to the 965 keV gamma ray, to see if any possible random sum pulses would affect the results.

The results of these measurements are summarized in Table 2.

It follows from above, especially 2) and 4), that the probability P'_K for K-capture decay to the 965 keV state was not determined in the same direct way as was P_K. P'_K is instead obtained from the relation

$$P'_K/P_K = (I_{\gamma x}/I_\gamma)_{965}/(I_{\gamma x}/I_\gamma)_{439},$$

Table 2.

$(I_{\gamma x})_{965}$	$(I_{\gamma})_{965}$	$(I_{\gamma x})_{439}$	$(I_{\gamma})_{439}$	$(I_{\gamma x}/I_{\gamma})_{965}/(I_{\gamma x}/I_{\gamma})_{439}$
560	53857	3116	198800	0.663 ± 0.039

deduced by the aid of formula (1). Inserting the experimental values, one finds $P'_K = 0.52 \pm 0.05$, which, according to formulae (2), corresponds to a decay energy of $145\ ^{+14}_{-10}$ keV. Combined with the gamma ray energy 965 ± 8 keV [11], this value gives a total decay energy

$$E_d(\mathrm{Tl}^{202}) = 1110 \pm 20 \text{ keV}.$$

The above value of the decay energy has been calculated on the assumption that the first and second excited states of the even-even nucleus Hg^{202} are both assigned 2+. The 2+ assignment of the second excited state is consistent with the nearly equal intensities found for the cross-over gamma ray of 965 keV and the cascade gamma ray of 523 keV.

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